Supplemental Material – Microscopic Origins of the Swim Pressure and the Anomalous Surface Tension of Active Matter

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I. LOCAL PRESSURE CALCULATION AND DISCUSSION

The local pressure – the flux of force across the surface S of a control volume V (cf. Fig. S1)– exerted by ABPs was computed and shown in Fig. 3C in the main text by making the following elemental arguments. From a thermodynamic perspective, the ideal Brownian osmotic pressure of colloids $\Pi^B = nk_BT$ is the result of the translational entropy of the particles. However, mechanically, this pressure manifests in different ways depending on the details of the particle dynamics. In the case of colloidal particles *with inertia* Π^B arises from the momenta of the particles with $\Pi^B = n\langle m\boldsymbol{U}\cdot\boldsymbol{U}/2\rangle$ where m is the mass of the particle and $\langle ...\rangle$ represents an average over all particles. From equipartition it follows that $\Pi^B = nk_BT$.

In the case of overdamped dynamics, the particle momentum is ill-defined and the Brownian osmotic pressure must be thought of us a *diffusive pressure*. To see this, we use the standard virial approach for computing the stress (e.g., taking the first spatial moment of the Brownian force) and find $\Pi^B = n\langle \boldsymbol{x}\cdot\boldsymbol{F^B}\rangle = n\zeta\int\langle \boldsymbol{U^B}(t')\cdot\boldsymbol{U^B}(t)\rangle dt$ where we now recognize $\int\langle \boldsymbol{U^B}(t')\cdot\boldsymbol{U^B}(t)\rangle dt$ as the particle's Brownian diffusivity D^B and $\Pi^B = n\zeta D^B$. The Brownian velocity is trivially related to the Brownian force $\boldsymbol{U^B} = \boldsymbol{F^B}/\zeta$ and is therefore also δ-correlated in time. This has the important implication that the Brownian osmotic pressure can be measured at any instant in time as the diffusivity can be instantaneously measured, in contrast to the swim diffusivity of active matter which requires a duration of τ_R before it can be measured [1].

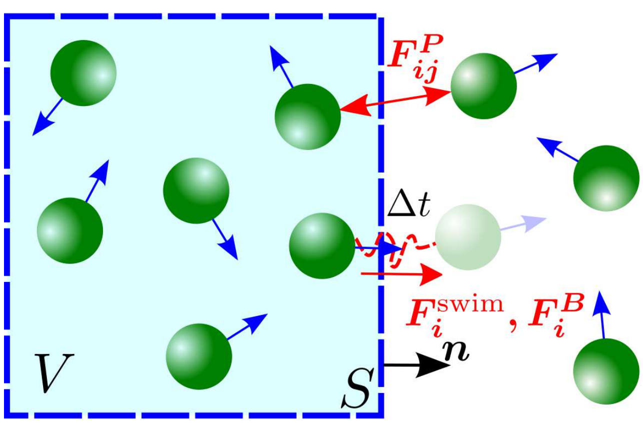

FIG. S1. Schematic for computing the local stress (flux of force across a plane).

Computing the local pressure of interacting particles with inertia can be readily achieved using the method-of-planes procedure [2] which, in a nutshell, computes the sum of interparticle force acting across a surface (see Fig. S1) and the rate of change of momentum in the control volume due to particles entering and exiting the surfaces. Importantly, the latter two fluxes can be measured *instantaneously* at the surface. We now propose an extension of the method-of-planes procedure to measure the local stresses generated by single-body forces, such as the Brownian and swim forces.

As a particle moves across a surface with a Brownian F^B and swim force $F^{\rm swim}$, it will exert a surface force on the particles in the neighboring control volume. We can compute this force by asking a simple question: what is the force required to keep the particles from crossing the imaginary surface plane? In other words, if the imaginary surface was an infinitely thin impenetrable wall, what force would the particles exert on the wall? This is precisely what we compute in Fig. 3 in the main text: at each simulate timestep we compute the force per unit area a hard wall would exert on the particles $\mathcal{F}$. The forces this hypothetical wall must exert to counteract the Brownian $\mathcal{F}^B$ and swim forces $\mathcal{F}^{\rm swim}$ can be readily distinguished (with $\mathcal{F} = \mathcal{F}^B + \mathcal{F}^{\rm swim}$). We note, however, that these quantities are not entirely decoupled as the frequency a particle crosses the surface is a function of both the Brownian and swim force. We further note that, as $\mathcal{F}$ should be interpreted as a local stress, its value is independent of which side of the hypothetical wall is used to measure it.

Single-body forces only contribute to surface-force flux *the instant* the particle is at the surface. It is for this reason that, when computing the swim force flux, we recover a local swim stress that is vanishingly small in comparison to the magnitude of the swim pressure. The swim diffusivity requires knowledge of the trajectory (the run length) of the particle. In contrast, the Brownian force at any instant in time fully encapsulates the Brownian diffusivity D_T of the particle and results in finding exactly the anticipated diffusive pressure $\mathcal{F}^B = n(z)\zeta D_T = n(z)k_BT$ at any instant in time and, hence, space, as shown in Fig. 3 in the main text.

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II. MOVIES

The file “dropletdynamics.mp4” contains a visualization of the dynamics of the active liquid droplet shown in Fig. 1A in the main text. The total duration of the video is $\approx 1100a/U_0$. “slabpreparation.mp4” illustrates our procedure for creating a single-liquid domain in the slab geometry.

[1] S. C. Takatori, W. Yan, and J. F. Brady, Phys. Rev. Lett. **113**, 028103 (2014).
[2] B. D. Todd, D. J. Evans, and P. J. Daivis, Phys. Rev. E **52**, 1627 (1995).